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Formation of a dielectric sublayer heterostructure of lead-tin telluride

A.K. Kaveev [1]✉

[1] Ioffe Institute, Saint-Petersburg, Russia
✉ kaveev@mail.ioffe.ru

Abstract. We have optimized the growth parameters of the buffer layer for further $Pb_{1-x}Sn_xTe$ ($x \geq 0.4$) deposition from the point of view of smoothness and crystalline quality. The latter has the properties of a crystalline topological insulator. A three-component heterostructure consisting of fluorite CaF_2, BaF_2, and cubic $Pb_{0.7}Sn_{0.3}Te$:In layers was formed on the Si(111). The surface morphology of this hybrid heterostructure was studied depending on the growth temperature and the thickness.

Keywords: Molecular beam epitaxy, $Pb_{0.7}Sn_{0.3}Te$, topological insulators

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Формирование диэлектрической гетероструктуры-подслоя для получения пленок теллурида свинца-олова

А.К. Кавеев [1]✉

[1] Физико-технический институт им. А.Ф. Иоффе РАН, Санкт-Петербург, Россия
✉ kaveev@mail.ioffe.ru

Аннотация. Оптимизированы ростовые параметры буферного подслоя для дальнейшего нанесения пленок $Pb_{1-x}Sn_xTe$ ($x \geq 0.4$) с точки зрения планарности и кристаллического качества. Трехкомпонентная гетероструктура, состоящая из слоев CaF_2, BaF_2, и $Pb_{0.7}Sn_{0.3}Te$:In сформирована на поверхности Si(111). Изучено влияние температуры роста и толщины на морфологию поверхности гетероструктуры.

Ключевые слова: молекулярно-лучевая эпитаксия, $Pb_{0.7}Sn_{0.3}Te$, топологические изоляторы

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Introduction

Crystalline topological insulators (TIs) based on the $Pb_{1-x}Sn_xTe$ compound are promising from the point of view of practical applications [1], due to the possibility of creating TIs with a volume less shunting the conductivity of topological states due to the possibility of controlling the composition (x value), in contrast to the already classical TIs based on V_2VI_3 compounds. In addition to controlling the composition, it is equally important to obtain low-defect $Pb_{1-x}Sn_xTe$ films,

also from the point of view of reducing the shunting effect of the volume. This task is non-trivial due to the problems that arise during the epitaxial growth of $Pb_{1-x}Sn_xTe$. To ensure the presence of conducting topological states, it is necessary to obtain a planar layer with a high crystalline quality.

Materials and Methods

CaF_2, BaF_2 and $Pb_{1-x}Sn_xTe$ films were deposited on Si(111) surface with use of conventional molecular beam epitaxy method, using a single crucible. Si mono-crystalline substrates were cleaned by Shiraki method [2] which represents sequential growth and etching of an oxide layer on the substrate. The final oxide layer was thermally eliminated in the growth chamber with formation of Si (111) 7×7 surface reconstruction. The crystallinity and epitaxial relations of the grown layers were controlled by in-situ reflection high energy electron diffraction (RHEED). The surface morphology was studied using atomic force microscopy technique in semi-contact mode.

Results and Discussion

In this work, optimization of the buffer layer for further deposition of $Pb_{1-x}Sn_xTe$ ($x \geq 0.4$), which has topological properties, was carried out. For this purpose, a multilayer structure consisting of CaF_2, BaF_2, and $Pb_{0.7}Sn_{0.3}Te$:In layers was deposited on the Si(111) surface. It is known that $BaF_2/CaF_2/Si(111)$ binary buffer layers were used for the epitaxial growth of $Pb_{1-x}Sn_xTe$ films on silicon [3]. At the same time, the height of the surface relief of the resulting buffer layers was too high due to insufficient optimization of the growth parameters and imperfection of these layers. It is known [4] that calcium fluoride, when applied to the (111) silicon surface by molecular beam epitaxy, forms layers of high crystalline quality, but different surface morphology, depending on the deposition temperature. The formation of islands of various heights and lateral sizes is possible. In this work, it was necessary to provide the smoothest possible surface. For this, the temperature regimes and the thickness of the calcium fluoride layer were varied. Next, a layer of barium fluoride was deposited on the surface of calcium fluoride, providing a smoother transition in the lattice constant from CaF_2 to $Pb_{1-x}Sn_xTe$. Figure 1, *a*–*g* shows a number of atomic force microscopy images of a $BaF_2/CaF_2/Si(111)$ heterostructure of various configurations. A number of parameters were determined that provide a combination of thin fluorite sublayers (which ensures their low imperfection) with their high smoothness. It has been shown that a decrease in the growth temperature of calcium fluoride and barium fluoride leads to a decrease in the lateral dimensions of the islands. The smoothest surface relief is achieved at low growth temperatures (250 °C). This correlates with the results of [5], where the high planarity of calcium fluoride films with a thickness of a unit of nanometers deposited at a given temperature was demonstrated. Attempts to increase the growth temperature of one of the two sublayers in order to improve the crystalline quality lead to an enlargement of the islands and coarsening of the relief, which is undesirable for the task of obtaining the smoothest layers posed in this work. It is also sometimes possible to form cracks in the film due to the difference in temperature expansion coefficients. At the same time, a fairly smooth relief is also obtained using two-stage (low and high temperatures) growth of calcium fluoride, followed by a low-temperature deposition of a layer of barium fluoride (Fig. 1, *e*). At the same time, direct deposition of a thin layer of $Pb_{1-x}Sn_xTe$ on a fluorite bilayer does not provide sufficient smoothness and sufficient crystalline quality for the formation of topological states. Good quality can be achieved only with the help of homoepitaxial growth of this material on a sufficiently thick sublayer. In this case, it is also necessary to achieve a low conductivity of this sublayer by doping it, which ensures the regulation of the position of the Fermi level. The experiments performed with fluorite bilayers made it possible to proceed to experiments with the deposition of the third buffer sublayer, the dielectric $Pb_{0.7}Sn_{0.3}Te$:In. This sublayer was grown at various temperatures from 250 to 450 °C (Fig. 1, *h*, and Fig. 2). The experiments also showed the importance of applying large (more than 500 nm) thicknesses of this sublayer to ensure its continuity, the absence of holes in the film, and to achieve a high surface planarity.

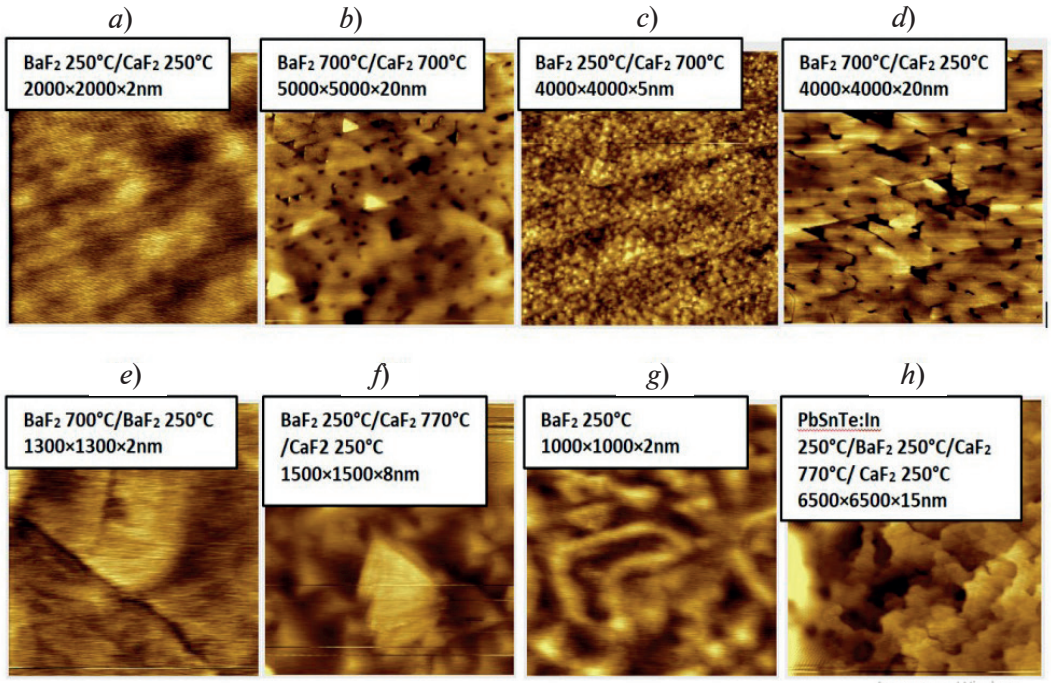

Fig. 1. Surface morphology of the BaF_2/CaF_2 buffer layer (*a*–*g*), grown under different conditions, and the optimized $Pb_{0.7}Sn_{0.3}Te$:In/BaF_2/CaF_2/Si(111) buffer layer (*h*). (*a*–*d*) BaF_2 and CaF_2 thickness is 20 and 10 nm, respectively. Layer thicknesses are 10 and 3 nm (*e*), 15, 7, and 3 nm (*f*), 20 nm (*g*), 300, 15, 7, and 3 nm (*h*)

Figure 2 shows examples of the surface morphology of the third buffer sublayer obtained under various conditions. The dielectric properties of this sublayer will be measured at the next stage of work by conducting electro-physical measurements. In the future, thin (10–20 nm) layers of stoichiometry close to $Pb_{0.6}Sn_{0.4}Te$ with an excess of Sn will be deposited on this sublayer, providing the presence of conducting topological states. The evolution of the surface during the formation of a three-layer buffer layer was studied in situ using RHEED technique.

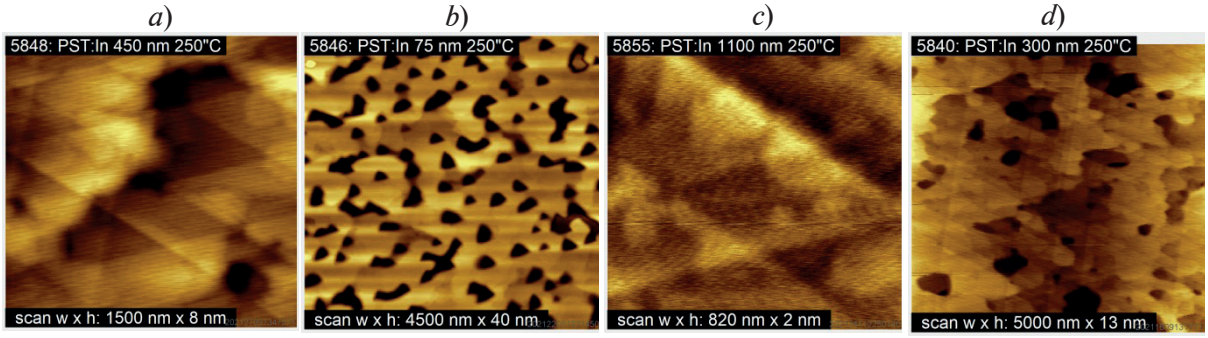

Fig. 2. Morphology of the surface of the third buffer sublayer Pb0.7Sn0.3Te:In depending on the growth conditions. (*a*) – deposition of BaF_2 and CaF_2 on the bilayer with parameters corresponding to Fig. 1, *f*; (*b*) – the same but thinner $Pb_{0.7}Sn_{0.3}Te$:In layer; (*c*) – deposition of BaF_2 and CaF_2 on the bilayer with parameters corresponding to Fig. 1, *a*; (*d*) – the same parameters, but a thinner $Pb_{0.7}Sn_{0.3}Te$:In layer grown a lower temperature

Figure 3 shows the change in RHEED patterns upon successive deposition of 10 nm calcium fluoride (*a*), 15 nm barium fluoride (*b*), and 1000 nm $Pb_{0.7}Sn_{0.3}Te$:In at the initial (*c*) and final (*d*) moment of growth at a temperature of 250 °C. It can be seen that calcium and barium fluorides grow in a planar manner, as evidenced by the streaks in the RHEED patterns. Further, at the initial stage of growth, the three-dimensional (island) growth of $Pb_{0.7}Sn_{0.3}Te$:In occurs.

Twinning of the reflections indicates a two-domain character of growth, which is characteristic of a cubic crystal lattice with a (111) growth surface. In our earlier work [6], trial experiments were carried out on the deposition of thin $Pb_{0.7}Sn_{0.3}Te$ films on Si(111) using a calcium fluoride buffer sublayer. Epitaxial relations with respect to silicon were established: (111) $Pb_{0.7}Sn_{0.3}Te \| (111)$ Si, $[1\bar{2}1]$ $Pb_{0.7}Sn_{0.3}Te \| [2\bar{1}\bar{1}]$ Si. In the present case, these relations are obviously preserved. With an increase in the thickness of the deposited material, point reflections are replaced by streaks, which indicate the fusion of islands (the Stranski-Krastanov growth regime) and the formation of a smooth surface. This RHEED pattern (Fig. 3, *d*) is in accordance with Fig. 2, *d*, which indeed demonstrates a smooth surface with characteristic dislocation glide plane exits at an angle of 60°, also characteristic of the growth surface (111) of the cubic crystal lattice.

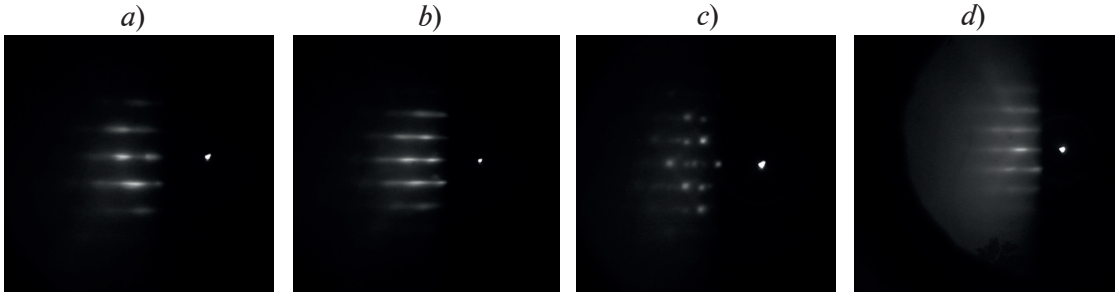

Fig. 3. Changes in RHEED patterns upon deposition of 10 nm calcium fluoride (*a*), 15 nm barium fluoride (*b*), and 1000 nm $Pb_{0.7}Sn_{0.3}Te$:In at initial (*c*) and final (*d*) moment of growth at a temperature of 250 °C

Conclusion

As a result of this work, a number of growth regimes were sorted out and growth parameters were optimized for the three-layer epitaxial heterostructure $Pb_{0.7}Sn_{0.3}Te$:In/BaF_2/CaF_2/Si(111). The possibility of planar low-defect growth of the third sublayer – $Pb_{0.7}Sn_{0.3}Te$:In, is shown, which is necessary for further homoepitaxial growth of $Pb_{1-x}Sn_xTe$ with an increased value of the parameter $x \geq 0.4$ in order to obtain topological surface states.

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THE AUTHORS

KAVEEV Andrey K. kaveev@mail.ioffe.ru

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